PROTON CAPTURE RESONANCES IN SULFUR

WILLIAM A. ALFONTE, JR.
and
DONALD C. TETTELBACH

PROTON CAPTURE RESONANCES IN SULFUR

* * * * *

William A. Alfonte, Jr.

and

Donald C. Tettelbach

PROTON CAPTURE

RESONANCES IN SULFUR

by

William A. Alfonte, Jr.
//
Captain United States Army

and

Donald C. Tettelbach

Lieutenant, United States Army

Submitted in partial fulfillment of
the requirements for the degree of

MASTER OF SCIENCE
IN
PHYSICS

United States Naval Postgraduate School
Monterey, California

1 9 6 1

PROTON CAPTURE RESONANCES IN SULFUR

by

William A. Alfonte, Jr.

and

Donald C. Tettelbach

This work is accepted as fulfilling
the thesis requirements for the degree of
MASTER OF SCIENCE
IN
PHYSICS
from the
United States Naval Postgraduate School

ABSTRACT

This investigation was conducted to identify resonances in the capture of protons by sulfur. Proton energies in the range between 1.15 and 1.30 Mev were utilized for this study.

Proton capture resonances which have not been previously reported were found at E_p of 1.165 and 1.269 Mev and attributed to S^{34}. The previously reported resonance of S^{33} at 1.264 and of S^{34} at 1.215 Mev (reported at 1.206 Mev) were confirmed. A possible S^{32} resonance was found at 1.302 Mev.

The authors wish to express their appreciation for the contributions of theoretical advice and assistance by Professor Edmund A. Milne, and for the technical assistance of K.C. Smith, M.C. Brillhardt, R. Moeller, and A. Goodall.

TABLE OF CONTENTS

Section	Title	Page
1.	Introduction	1
2.	Resonance and Decay Phenomena	5
3.	Previous Investigations	7
4.	Equipment	8
5.	Target Preparation	11
6.	Experimental Procedures	13
7.	Results and Conclusions	15
	Bibliography	17
	Table I	18
	Figures 5 - 8	19

LIST OF ILLUSTRATIONS

Figure		Page
1.	Energy Levels and Decay Scheme for Cl^{33}	2
2.	Energy Levels and Decay Scheme for Cl^{34}	3
3.	Energy Levels and Decay Scheme for Cl^{35}	4
4.	Energy Levels and Decay Scheme for Cl^{37}	4
5.	Magnet Current Calibration Curve	19
6.	Target and Mount Assembly	20
7.	Equipment Block Diagram	21
8.	Photograph of Target Assembly	22

1. Introduction.

This investigation was conducted to extend the previous work done at the US Naval Postgraduate School on the determination of resonances in the capture of protons by sulfur. Proton energies between 1.15 and 1.30 Mev were studied. Improved equipment with better resolution and identification characteristics was used.

Naturally-occurring sulfur is composed of four isotopes as follows:

ISOTOPE	ABUNDANCE
S^{32}	95.1%
S^{33}	0.74%
S^{34}	4.2%
S^{36}	0.016%

Bombardment of these sulfur nuclides with protons results in the formation of compound nuclides of chlorine. The compound nuclides $[Cl^{33}]$ and $[Cl^{34}]$, formed from S^{32} and S^{33} respectively, decay by particle re-emission (elastic or inelastic scattering) or gamma ray emission. The ground states of Cl^{33} and Cl^{34} decay by positron emission with decay schemes as shown in Figures 1 and 2. The compound nuclides $[Cl^{35}]$ and $[Cl^{37}]$ are formed by the similar proton reactions of the two sulfur isotopes S^{34} and S^{36} and also decay by particle re-emission or by gamma ray emission to their ground states as shown in Figures 3 and 4.

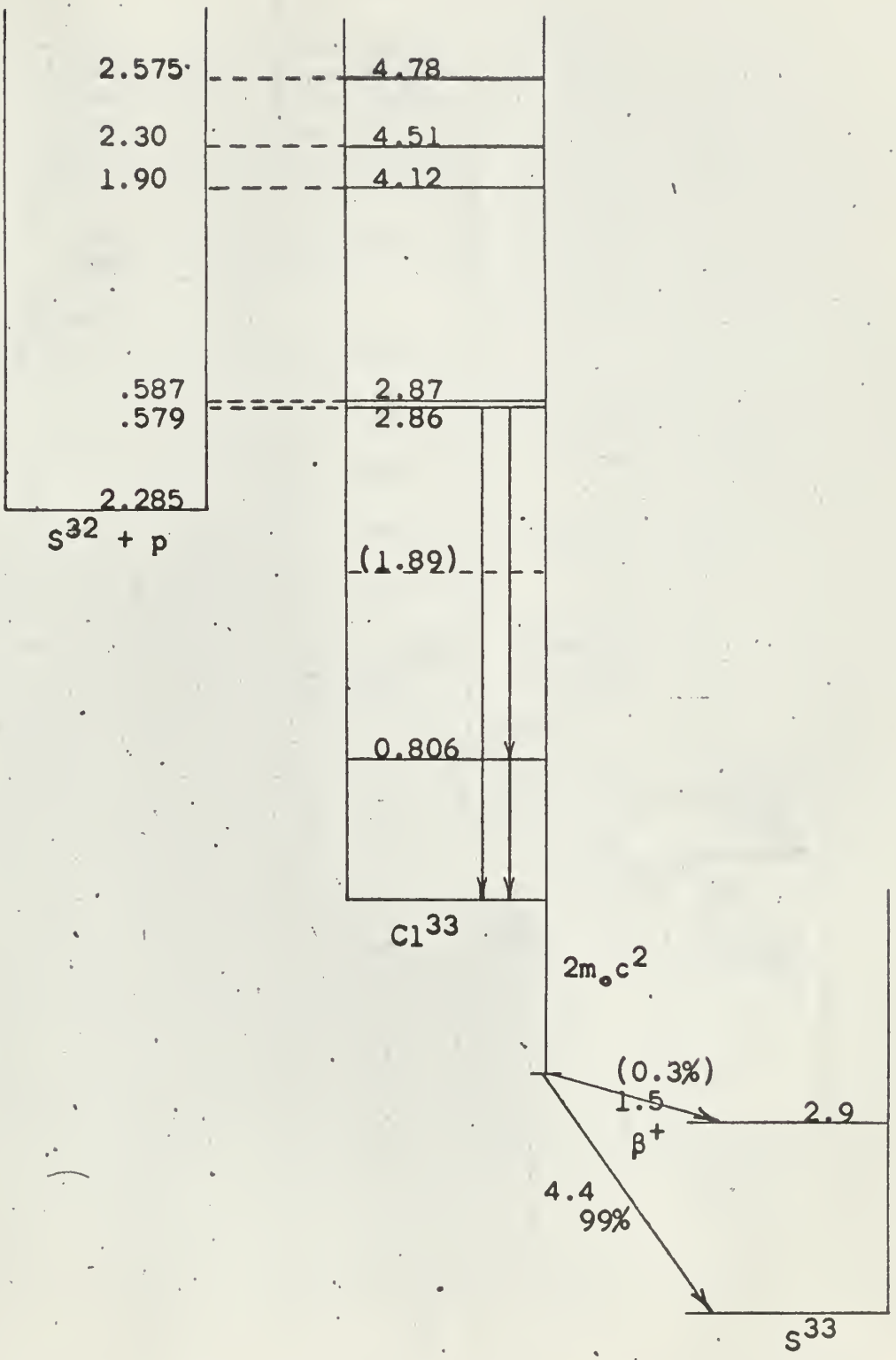


Figure 1. Energy Levels and Decay Scheme of Cl^{33}

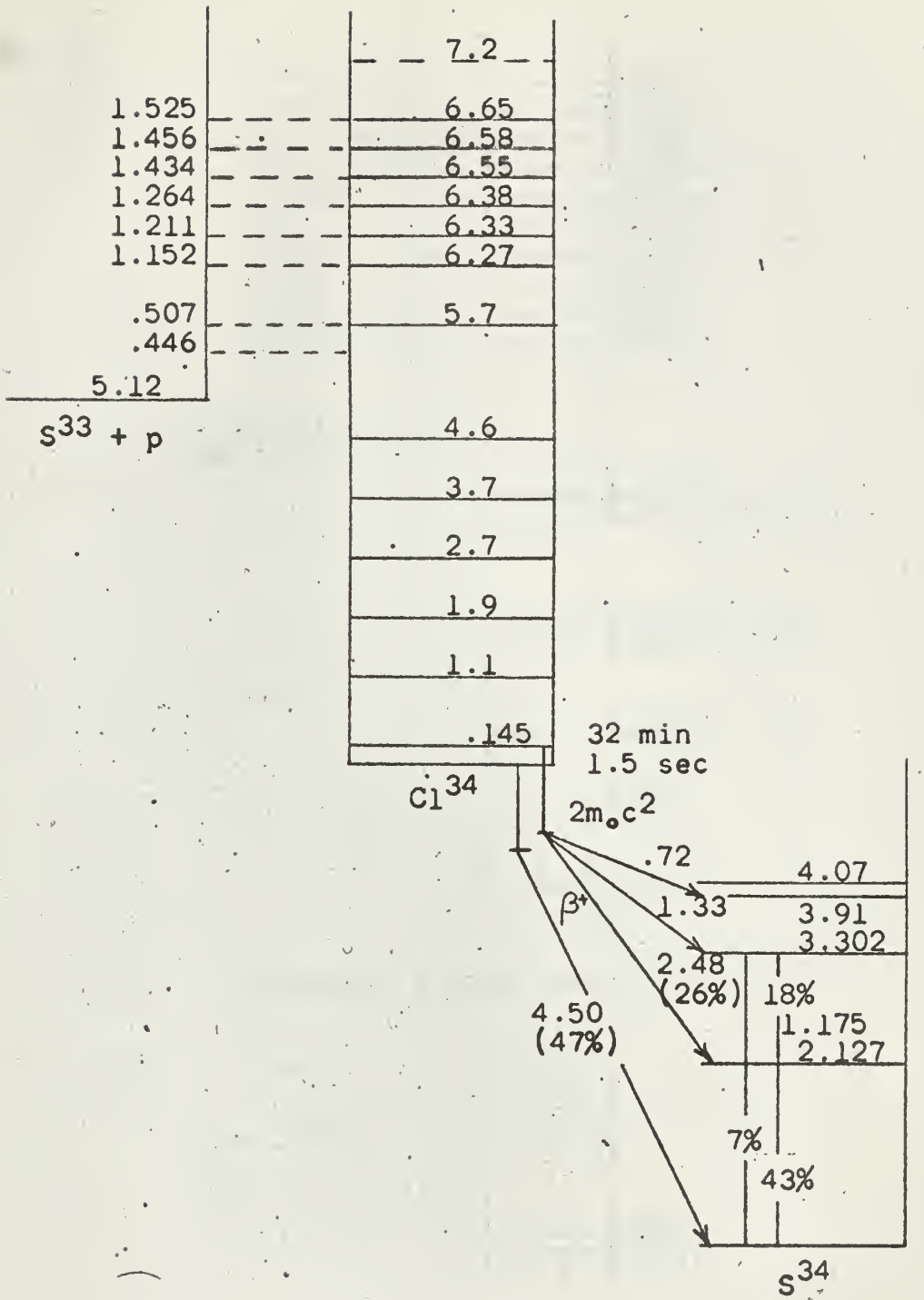


Figure 2. Energy Levels and Decay Scheme of Cl^{34}

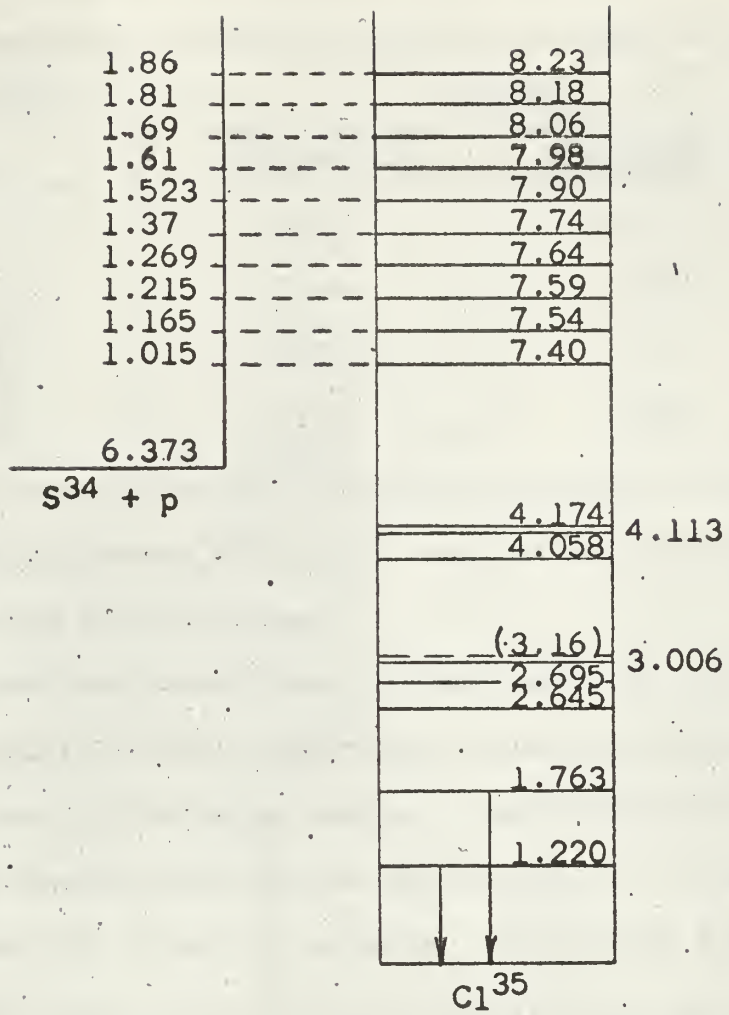


Figure 3. Energy Levels in Cl^{35}

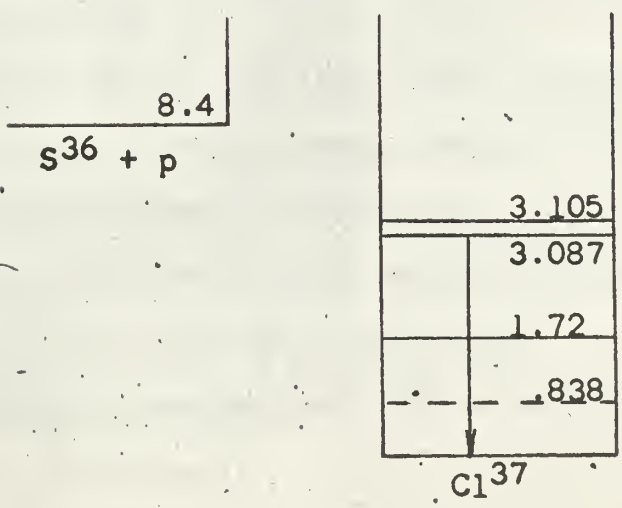


Figure 4. Energy Levels in Cl^{37}

To facilitate the study, samples of cadmium sulfide enriched in S^{33} and S^{34} were vacuum coated on 10 mil tantalum target discs. These enriched samples had the following composition by weight of sulfur:

ISOTOPE	TARGET	
	ENRICHED S^{33}	ENRICHED S^{34}
S^{32}	73.6%	62%
S^{33}	22.1%	0.8%
S^{34}	4.2%	37.2%
S^{36}	-	TRACE

The low concentration of S^{36} in both the natural and enriched sulfur samples indicate that the $S^{36}(p,\gamma)Cl^{37}$ reaction could be safely neglected.

2. Resonance and Decay Phenomena

In a transmutation experiment of light nuclei it is believed that a compound nucleus is formed in the initial interaction between the bombarding particle and the target nucleus. The characteristics of the compound nucleus are dependent upon the proton-neutron configuration and are independent of its mode of formation. The compound nucleus does display discrete energy states and therefore the accurate determination of the bombarding particle energy resulting in formation of a compound nucleus is of interest.

The compound nucleus will disintegrate in approximately 10^{-14} seconds in accordance with the energy available and the selection rules. Some of the possible modes of interest are:

a) re-emission of the incident particle with the same or lower energy (elastic or inelastic scattering)

b) emission of other particles such as neutrons, deuterons, alpha particles, or protons.

c) gamma radiation to the ground state or to an intermediate energy state of the compound nucleus.

The residual nucleus remaining after one of the above processes may be in an excited state, but is usually stable with respect to heavy particle emission. If it is in an excited state, gamma radiation results, returning the residual nucleus to its ground state.

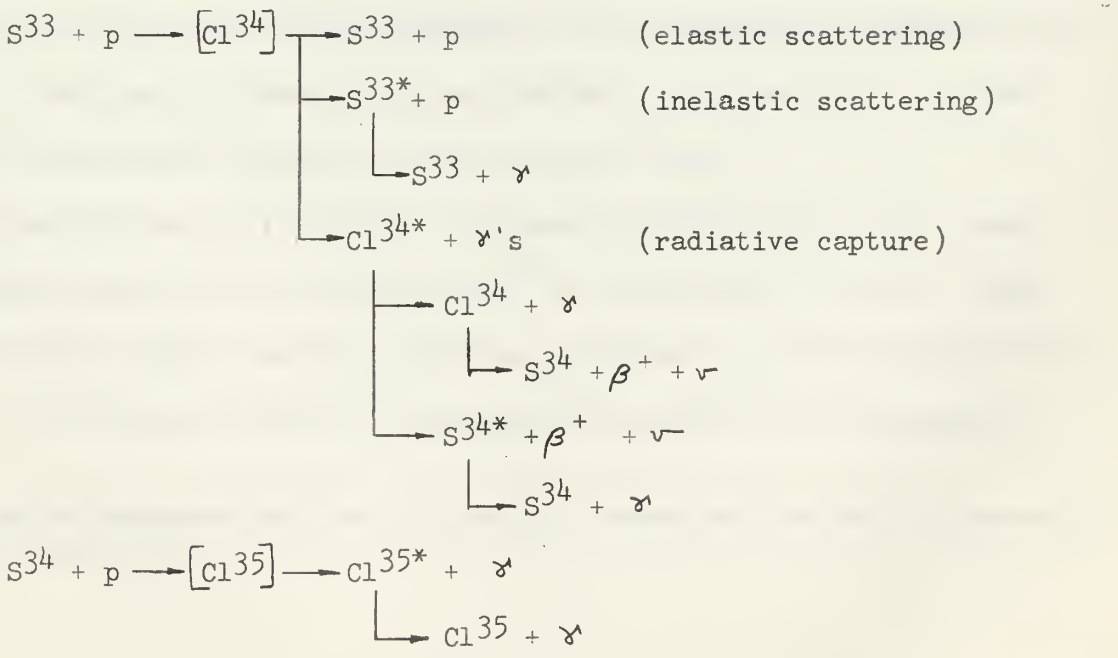
The energy of the gamma radiation is dependent upon the relative energy of the two nuclear states involved.

$$h\nu = E_1 - E_2$$

where E_1 represents the energy of the initial state and E_2 represents the energy of the final state.

Thus if E_1 is dependent upon the energy of the incident particle, as it is in the compound nucleus, the energy of the gamma is dependent upon the energy of the incident particle. However, if E_1 represents an energy level in a residual nucleus, the gamma ray energy is independent of the energy of the bombarding particle.

In this investigation the types of reactions of interest were:



A more comprehensive treatment of this subject has been published by Fowler et al [1].

3. Previous Investigations

Hanscome [2] and Malich at the Naval Research Laboratories had investigated the S(p,γ)Cl reactions in the proton energy range of 0.9 to 2.1 Mev prior to 1951. Resonances were observed at proton energies[1] of about 1.37, 1.61, 1.69, 1.81 and 1.86 Mev and all were attributed to the isotope S^{34}.

Ferguson [3] and Gove at the Chalk River Laboratories conducted investigations in the proton energy (E_p) range of 1.0 to 2.8 Mev. Resonances were found at 1.9 and 2.31 Mev and attributed to S^{32}.

More recently Olness [4] et al, working at Duke University, studied the energy range between 1.5 and 4.0 Mev. Their work confirmed the resonances reported at 1.9 and 2.31 Mev for the isotope S^{32}.

C. Van der Leun [5] in 1958 reported a quite comprehensive study of the (p,γ) reactions of several of the light elements, including the isotopes S^{32} and S^{33}. His experiments concerned protons with energies in the range up to 800 kev and included two S^{33} resonances at 449 and 513 kev. Subsequently, Kuperus [6] and Smulders, collaborating with Endt, have revised these figures to 446.5 and 507.1 kev.

Van der Leun [7] and Endt confirmed by Kuperus [6] et al, have further reported two resonances in S^{32} at 579.8 and 587.4 kev. These replace the single resonance previously reported at 594 kev by Endt [8]. These are the only known S^{32} resonances below the 1.9 Mev resonance.

[1]Proton energies reported in previous investigations are in laboratory coordinate system.

In 1959 Gaehler [9] and Knipp investigated the isotope S^{33} in the proton energy range between 1.10 and 1.55 Mev. Resonances were reported at 1.152, 1.211, 1.262, 1.434, 1.456, and 1.525 Mev.

Concurrently, Moore [10] and Krumwiede were investigating the isotope S^{34} in the proton energy range of 800 kev to 1.95 Mev. Resonances were reported at E_p of 1.015, 1.206, and 1.523 Mev. Three additional possible resonances were indicated but the presence of fluorine contamination precluded positive identification.

A tabulation of these results appears in TABLE I.

4. Equipment

The 2-Mev Van de Graaff electrostatic accelerator at the US Naval Postgraduate School was used to accelerate the protons necessary for this investigation. A 25° magnetic analyzer (momentum selector) was used in conjunction with a pair of adjustable slits 2.1 meters from the center of the magnet to obtain the desired beam of mono-energetic protons. Selection of the proper magnet current provided a proton beam of the desired energy. The magnet current was measured with a Leeds and Northrup potentiometer with a relative accuracy of 2×10^{-4} amperes, corresponding to a proton energy of approximately 0.3 Kev. The slits refined the energy spread of the proton beam for increased energy resolution. Resolution of 0.2% [13] was obtainable for a 1-Mev proton beam with the slit width adjusted to 8×10^{-4} meters.

A current integrator was used to measure the total accumulated charge on the target as a result of proton bombardment. This was done by transferring the charge to a capacitor until a fixed voltage was obtained. This voltage activated a stop relay in the current integrator.

By changing the size of the capacitor, the charge required, and therefore the quantity of proton bombardment, was changed.

The stop relay in the current integrator serves three primary functions:

a) controls a beam shutter between the viewer slit control box and the target which blocks the proton beam from the target.

b) controls a TIME scalar which provides a means of recording the actual period of time the target is bombarded.

c) controls the COUNT scalar. (See page 10)

The targets were mounted on a vacuum-tight turret capable of holding four target mounts which could be interchanged without disturbing the vacuum. Two types of target mounts were utilized: 1) 10mm glass tubing (1.3 cm OD) three inches long, held in the target turret by a glass to metal quick vacuum coupling; 2) an extension on the above target mount of 8 mm glass tubing (1.0 cm OD) vacuum sealed and connected with vacuum hose, Figure 6. The tantalum target discs were held on by three methods: 1) vacuum pressure using a gasket of surgical rubber; 2) vacuum pressure with O ring gasket; 3) Glyptal cement. Methods 1 and 2 provided a capability for a rapid change of targets without changing the target mounts. The surgical rubber gasket melted on occasion during prolonged bombardment and caused a partial loss of vacuum. The use of glyptal cement required the preparation of the targets and mounts a week in advance and introduced occasional vacuum leaks. Two targets with extensions were placed on opposite sides of the turret for use in the well of a four-inch scintillation crystal while the other two without extensions could be used outside the well.

To prevent the electrons escaping the target from being accelerated to the high potential dome, a 300-volt negative retarding potential was placed on a brass ring about 4 cm from the target end of the 10-cm mounts. A 300-volt battery connected through a 430 K ohm resistor provided the negative potential.

Cooling of the targets was accomplished by directing dry nitrogen on the targets after passing it through a coil in a liquid air bath. Glass tubing was bent to run alongside the target mounts when placed inside the crystal well to carry the cooling nitrogen and the current integrator lead to the target.

The gamma yield was detected by the 4 inch thallium-activated sodium iodide crystal mounted on a Dumont 6354 photomultiplier tube. The signals from the phototube passed through a cathode follower to a non-overloading amplifier and discriminator to the scalar controlled by the current integrator.

An RCLaic 128 Channel Analyzer was simultaneously connected to the pre-amplifier of the photomultiplier tube to identify the gamma spectrum. The 128 channel analyzer, which also supplied power to the phototube, was modified to use either an external high voltage supply or its internal high voltage supply. This was done to stabilize the high voltage for the analyzer when analyzing the gamma ray spectrum as the internal power supply of the analyzer did not demonstrate the desired stability. The internal power supply was suitable to maintain the proper voltage for use of the phototube with the scalar. The power supplies could be selected by means of a switch on the 128 channel analyzer.

The crystal and phototube were shielded from stray background radiation by constructing a lead pig enclosing the entire target area. The phototube and crystal were mounted on tracks to allow easy and accurate positioning of the crystal without damaging the targets.

To make it easier to make small changes in the magnet current, the helipot for the magnet current control was also modified by connecting a 5K ohm resistor in series and a 10K ohm resistor in parallel with the helipot.

5. Target Preparation

The difficulties reported by Moore and Krumwiede [10] in the use of cadmium sulfide as a target material, together with the limited amount of enriched CdS^{33} available, made it necessary to investigate the properties of other sulfur compounds for use as target materials. Limited additional quantities of enriched sulfur isotopes in the form of pure sulfur were available for use.

To preclude any significant proton reactions with materials on the target other than the sulfur it was considered that heavy metal sulfides would be the most desirable type of compound. However, since some enriched cadmium sulfide isotopes were available the initial attempts at target preparation were made with natural cadmium sulfide.

Discs were stamped out of 10-mil tantalum sheet for use as target backing. Extreme care was used constantly while manipulating target materials to prevent contamination with foreign materials.

The method described by Moore and Krumwiede for the preparation of CdS targets was tried with only limited success. It appeared that a vacuum coating system with two separate cold traps would be more feasible,

with one cold trap providing the desired working vacuum and the second trap for mounting the targets to permit target cooling during evaporation of the CdS.

Such a system was used to coat targets at a vacuum better that 10^{-6} mm Hg.[1] The target discs were cleaned with nitric acid and acetone and mounted on the bottom of the cold trap directly over the boat containing the material to be evaporated. The desired weight of CdS was placed in the boat. The CdS was weighed on a Christian Becker balance. The system was evacuated and, when a pressure of about 4×10^{-6} mm Hg was indicated by the VG1A ionization guage, the main cold trap was cooled with liquid air. In about ten minutes the cold trap with the targets mounted was cooled with liquid air and the CdS sample evaporated.

Calculations based on the area coated indicated that 1 mg of CdS would give a coating density on the targets of 20 microgram (μg) per cm^2. Targets were prepared with densities of 40 and 60 μg per cm^2.

Concurrently, the use of the sulfides of silver and copper was being evaluated. Target discs, after cleaning with nitric acid and acetone, were vacuum coated with either metallic silver or copper. These materials could be readily coated by conventional vacuum coating techniques and this was accomplished at a pressure of about 2×10^{-5} mm Hg. These "plated" targets were then treated in a solution of H_2S prepared by bubbling H_2S gas through distilled water. This resulted in formation of a layer of sulfide on the surface of the target, the density of which could be estimated visually and the reaction terminated by removing the targets from

[1]The system utilized was described by Fong & Jacobs in Thesis, USNPGS, "Superconductivity Properties of Evaporated Thin Tin Films with Metallic Backing" (1959).

the solution and rinsing with distilled water. This generally resulted in a coating which appeared uneven, but which, upon subsequent bombardment, resulted in quite sharp resonances, and showed no evidence of contamination.

Also, silver sulfide was prepared by passing H_2S gas through a solution of silver nitrate and the separation of the resultant precipitate. This was vacuum coated on target discs cleaned with acetone at a pressure of 2×10^{-5} mm Hg. The targets were not cooled during the evaporation phase of the operation. Visual inspection of the coated targets indicated a very limited amount of the AgS had "stuck". This was confirmed subsequently by the very small but detectable resonances indicated during proton bombardment.

Attempts to generate H_2S gas using about 5 mg of natural sulfur were unsuccessful. When bubbled into a solution of silver nitrate, only a minute amount of precipitate was formed indicating essentially a complete loss of the sulfur.

Since only 5 mg of enriched S^{33} were available and no technique to convert this amount to a sulfide was developed, the method of target preparation selected was to use the available enriched CdS, employing the improved coating techniques of the two cold trap system.

6. Experimental Procedure

Prior to investigating S^{33} and S^{34} the magnetic analyzer of the Van de Graaff accelerator was calibrated for proton energy versus magnet

current using known lithium fluoride resonances. The following resonances [11] were used to determine the calibration curve.

$F^{19}(p,\alpha)O^{16}$	(E_p Mev, Assumed 0.2% accuracy)	$Li^7(p,n)Be^7$
0.8735 ± 0.0008	1.283 ± 0.003	(Threshold)
0.935 ± 0.002	1.346 ± 0.003	1.8811 ± 0.0015
1.092 ± 0.002	1.372 ± 0.003	
1.137 ± 0.002	1.670 ± 0.010	
1.176 ± 0.002		

The proton energy range from 1.15 Mev to 1.30 Mev was traversed with the Van de Graaff accelerator in increments of 0.0010 amperes of magnet current corresponding to proton energy increments of approximately 1.6 Kev. The amplifier pulse height discriminator was biased at 1.5 Mev to eliminate all possible low energy background counting. The time, number of counts, and magnet current were recorded at each increment and background counts were made every fifth increment. A mean background count rate for the inclosed five increments was calculated and subtracted from each increment according to the time of the run. A plot was made of counts versus energy and possible resonances were determined and recorded for closer investigation.

Possible sulfur resonances were then investigated by again traversing the proton energy range in smaller magnet current increments in the region of a suspected sulfur resonance. Both enriched CdS^{33} and CdS^{34} targets were investigated as well as a natural CuS target. Calibration was achieved by observing the known $F^{19}(p,\alpha)O^{16}$ resonances at 1.092 and 1.137 Mev initially and 1.346 and 1.372 Mev after traversing the proton energy range. Each resonance was further studied with the RCLaic 128 Channel Analyzer to obtain a gamma ray spectrum.

The proton energy of the sulfur resonances was determined directly by entering the calibration curve with a corrected magnet current. This was necessary since it was found that linear extrapolation of magnet current from a calibration magnet current to an unknown resonance greater than 1×10^{-2} amperes resulted in rapidly increasing the uncertainty of the proton energy.

For each run the initial and final calibration magnet currents were compared to the calibration curve to compute a correction factor. This factor, when applied to an observed magnet current reading, gave the correct magnet current to enter the calibration curve.

The gamma spectra of previously reported S^{33} and S^{34} resonances and suspected contaminating resonances of fluorine and carbon were analyzed with the RCLaic 128 Channel Analyzer. The unknown sulfur resonances were then identified by comparison of their gamma ray spectra with the known spectra.

7. Results and Conclusions

a) A resonance in S^{34} was identified at a proton energy of 1.165 $\pm$ 0.004 Mev. This resonance was partially masked by the 1.176 Mev fluorine resonance but was clearly demonstrated by the relative magnitude of the gamma ray yield from the CdS^{34} target when compared with those of CdS^{33} and CuS , and by the gamma ray spectra from these three targets. The gamma ray spectrum of the CuS target was especially informative, being relatively free of fluorine contamination.

b) The S^{34} resonance previously reported at a proton energy of 1.206 Mev was identified at 1.215 $\pm$ 0.004 Mev.

c) Small resonances occurred on both sides of the 1.215 S^{34} resonance. These were located at 1.205 and 1.220 $\pm$ 0.004 Mev but positive

identification was not made. It is assumed that one of these corresponds to the S^{33} resonance reported at 1.211 Mev.

d) A resonance in S^{34} at a proton energy of 1.269 $\pm$ 0.004 Mev was identified based on the comparison of the rise found on the CdS^{34} target with that of the natural CuS. Also the CdS^{33} targets showed two peaks in this region, one corresponding to the S^{34} resonance and one to the 1.283 Mev fluorine resonance. The gamma ray spectrum at various portions of this peak corroborate the identification.

e) The resonance reported by Gaehler and Knipp at a proton energy of 1.262 Mev was confirmed as being S^{33} and located at 1.264 $\pm$ 0.004 Mev.

f) A resonance was detected at 1.152 $\pm$ 0.004 Mev but was not identified.

g) A resonance was found at a proton energy of 1.302 $\pm$ 0.004 Mev. Although not identified, this resonance is possibly due to S^{32} since it was much more prominent on the natural CuS targets than on either CdS^{33} or CdS^{34}.

BIBLIOGRAPHY

1. W. A. Fowler, C. C. Lauritsen, & T. Lauritsen "Gamma Radiation from Excited States of Light Nuclei". Rev Mod Phys 20, 236, (1948).

2. T. D. Hanscome & C. W. Malich, "The Reactions S(p,γ)Cl". Phys Rev, 82, 30 (A), (1951).

3. A. J. Ferguson & H. E. Gove, "Resonances in $S^{32}(p,p)S^{32}$". Phys Rev, 91, 439A, (1953).

4. J. W. Olness, W. Haeberli, & H. W. Lewis, "Levels of Cl^{33} from $S^{32}(p,p)S^{32}(p,p',\gamma)S^{32}$". Phys Rev, 112, 1702, (1958).

5. C. Van der Leun, Thesis "Investigations with Light Nuclei with (p,γ) Reactions". Utrecht University, Utrecht, Nederland (1958).

6. J. Kuperus, P. J. M. Smulders & P. M. Endt, "Energy Measurements of Proton Resonances in Light Nuclei". Physica, 25, 600, (1959).

7. C. Van der Leun & P. M. Endt, "Doublet Resonance in the Reaction $S^{32}(p,\gamma)Cl^{33}$". Physica 24, 1095, (1958).

8. P. M. Endt, "The Reaction $S^{32}(p,\gamma)Cl^{33}$". Physica 22, 1234, (Dec 1956).

9. A. H. Gaehler & A. L. Knipp, Jr., Thesis "Resonances in the Capture of Protons by Sulfur 33". USNPGS (1959).

10. J. A. Moore & J. L. Krumwiede, Thesis "Resonances in the Radiative Capture of Protons by Sulfur 34". USNPGS (1959).

11. F. Ajzenberg & T. Lauritsen, "Energy Levels of Light Nuclei". Rev Mod Phys, 27, 77, (1955).

12. D. Strominger, J. M. Hollander, & G. T. Seaborg, "Table of Isotopes". Rev Mod Phys, 30, 585, (1958).

13. J. T. Lewis & C. J. Reidel, Thesis "Energy Levels in P^{30}". USNPGS (1960).

TABLE I

RESULTS OF PREVIOUS INVESTIGATIONS OF PROTON RESONANCES IN SULFUR

S^{32}	S^{33}	S^{34}
	446.5±3.5 (1)	
	507.1±1.0 (1)	
579.8±1.1 (1)		
587.3±1.1 (1)		
		1015.±4. (2)
	1152.±2. (3)	
		1206.±4. (2)
	1211.±2. (3)	
	1262.±2. (3)	
		~1370 (4)
	1434.±2. (3)	
	1456.±2. (3)	
		1523.±4. (2)
	1525.±2. (3)	
		~1610 (4)
		~1690 (4)
		~1810 (4)
		~1860 (4)
1900.±2. (5)		
2310.±4. (5)		

(1) J. Kuperus
(2) Moore & Krumweide
(3) Gaehler & Knipp
(4) Hanscome & Malich
(5) Olness, Haeberli, & Lewis

Energies in kev., laboratory coordinate system.

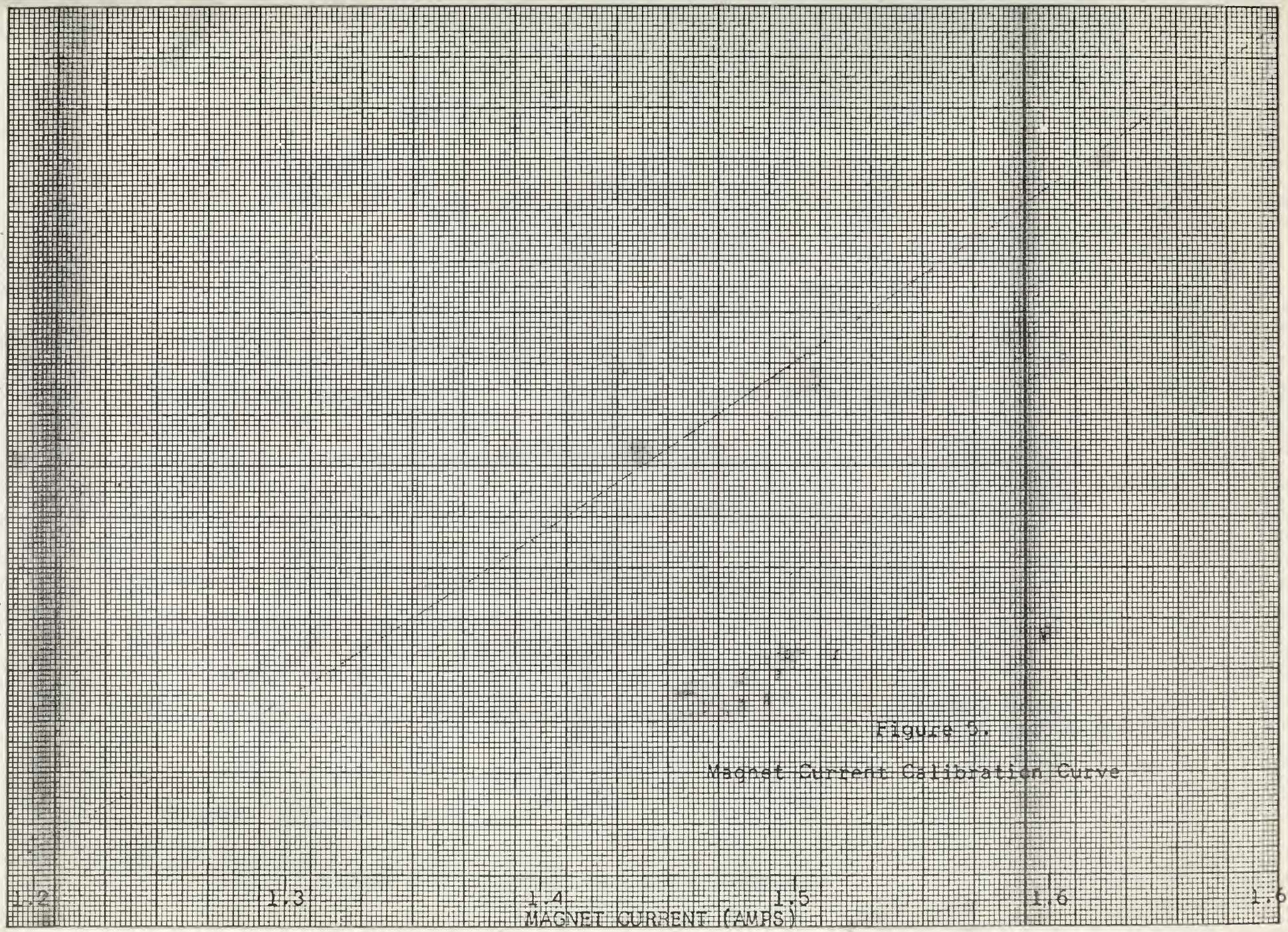


Figure 5.

Magnet Current Calibration Curve

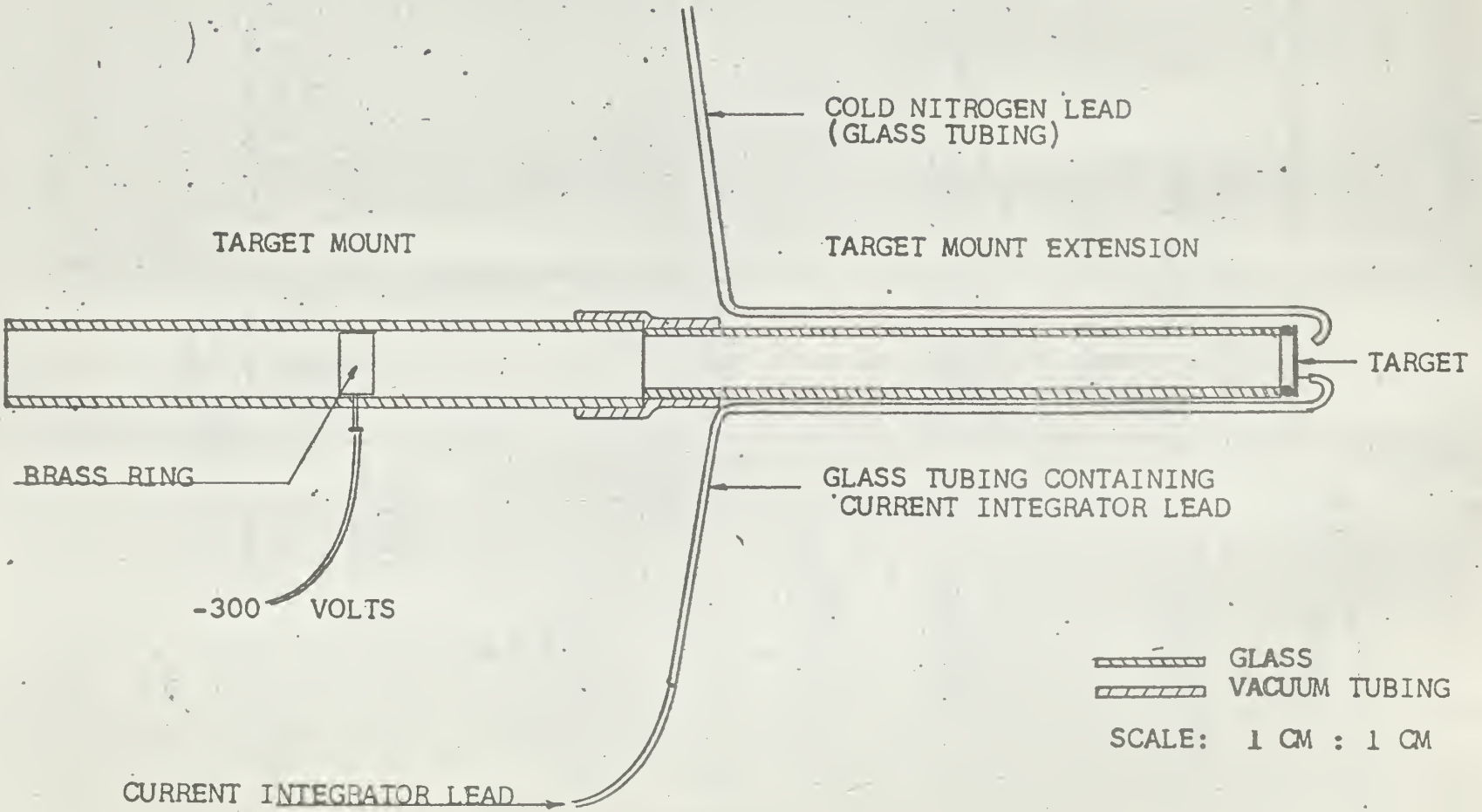


FIGURE 6. TARGET AND MOUNT ASSEMBLY

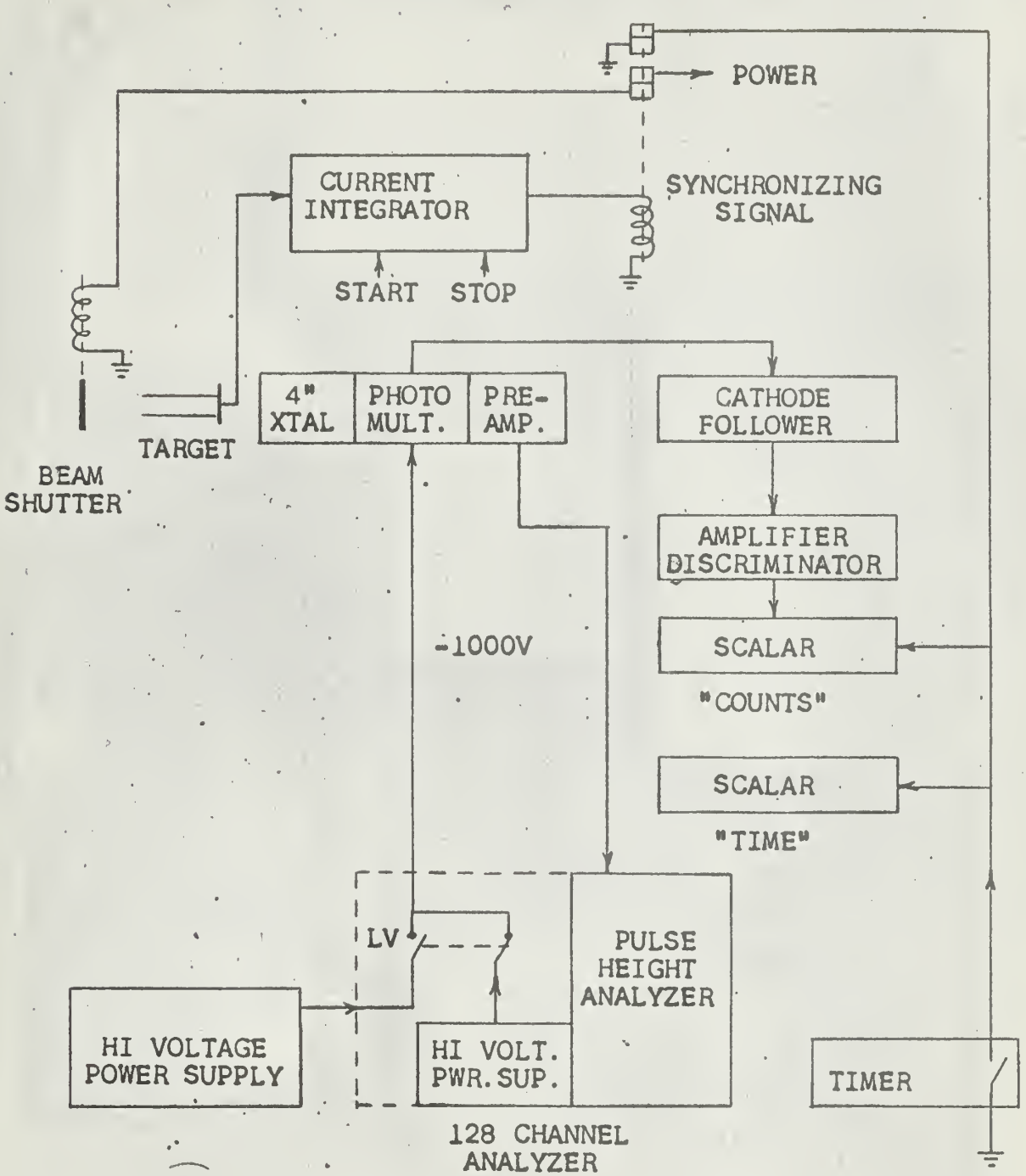
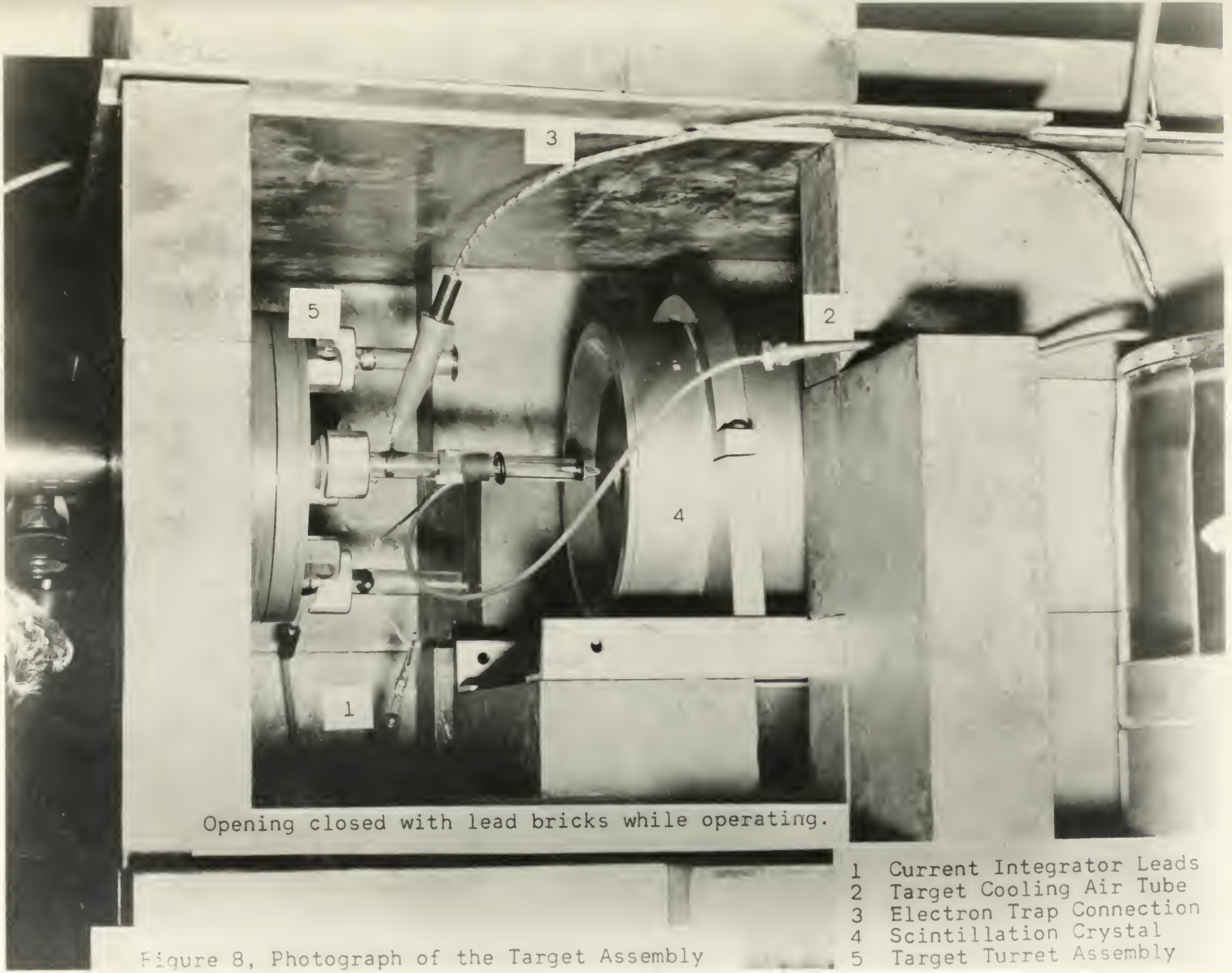


Figure 7. Equipment Block Diagram



Opening closed with lead bricks while operating.

1 Current Integrator Leads
2 Target Cooling Air Tube
3 Electron Trap Connection
4 Scintillation Crystal
5 Target Turret Assembly

Figure 8, Photograph of the Target Assembly